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Two-photon and EIT-assisted Doppler cooling in a three-level cascade system

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Laser cooling processes are theoretically investigated for a cascade scheme of atomic levels where the upper state decays more slowly than the intermediate one. A laser coupling to the upper transition modifies the scattering cross section, such that its action results in temperatures lower than those reached by Doppler cooling on the lower levels. We identify two regimes: when multiphoton processes due to the upper laser are relevant, the formation of an atomic coherence between ground and upper state affects the cooling dynamics, and the final temperature is controlled by the second laser parameters. When the intermediate state is only virtually excited, the dynamics are dominated by the two-photon process and the final temperature is determined by the spontaneous decay rate of the upper state.

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Ultracold temperatures in atomic gases are reached by means of laser cooling [1], evaporative cooling [2], sympathetic cooling [3] or stochastic cooling[4]. In order to decrease the kinetic energy associated with the atomic center-of-mass motion, laser cooling is based on the exchange of momentum between laser light and a closed atomic (or molecular) system consisting of a few active levels. In Doppler cooling the action of light scattering on the atomic motion can be described by means of a force [5]. The basic ingredients are i) an atomic cross section with a resonance enhancing the photon absorption and the force acting on the atoms; ii) a dependence of the absorption process on the atomic momentum leading to a dependence of the force on the momentum. If the resulting force damps the atomic momentum, the cooling process compresses the atomic momenta into a narrow distribution, from which one extracts the laser cooling temperature.

Laser cooling techniques have been demonstrated to be very efficient for alkali atoms, where temperatures down to several hundreds of nanoKelvins have been reached [1]. Much lower efficiencies have been achieved for other atomic systems, and in particular for alkaline-earth-metal atoms. One attractive feature of group-II elements is the simple internal structure with no hyperfine levels for the most abundant bosonic isotopes, which make them particularly attractive for improved frequency standards and optical clocks. However because the ground state is non-degenerate, sub-Doppler cooling is not possible and the temperature for Doppler cooling on the resonance line is typically limited to a few milliKelvins. For these elements other cooling strategies have been employed, such as cooling on the intercombination singlet-triplet line, as pioneered in [6, 7], or quench cooling with the lifetime of a triplet state modified by the resonant coupling to a fast decaying singlet state, as tested on calcium[8, 9].

An alternative cooling strategy for alkaline-earth atoms employs a two-color laser excitation of a three-level cascade configuration and uses the atomic coherence created between the terminal states of the cascade. This scheme was initially explored on metastable helium with limited efficiency[10, 11]. Theoretical analyses for alkali-earth and ytterbium atoms were presented in [12, 13, 14]. Experimental evidence of improved laser cooling on the cascade was presented for magnesium atoms in [15, 16]. This cooling strategy is based on mixing by laser radiation the excited state of the fast decaying singlet state with another state having a longer lifetime. Because the Doppler-cooling temperature depends inversely on the excited state lifetime, a lower temperature is produced by the excited state mixing. The scheme has flexible handles in the frequency and intensity of the mixing laser.

The present work analyzes theoretically laser cooling on a cascade configuration of levels, where a stable atomic level (ground state) is coupled by a laser to an excited level (intermediate state), which itself is coupled by a second laser to a higher energy state (upper state). The intermediate state decays more rapidly than the upper state, and the laser coupling to the upper transition modifies the absorption on the lower transition. This configuration is found in group-II elements (see the first column in Table I) and it supports the creation of a stationary atomic coherence between the ground and upper state, as introduced in [17]. Depending on the role of the intermediate state in the scattering processes we identify two different regimes, which characterize the cooling dynamics. In the first one the formation of atomic coherence between the ground and the upper state critically affects the properties of the force. We denote this regime as cooling assisted by electromagnetic-induced transparency (EIT), similar to that realized in a lambda configuration [18]. In the EIT-assisted cooling a temperature lower than the Doppler limit on the lower transition is achieved. The

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TABLE I: Parameters for cascade atomic transitions.

Parameter	Mg	Ca	Cs
1^{st} Trans.	$3^1S_0 \to 3^1P_1$	$4^1S_0 \to 4^1P_1$	$6^2S_{1/2} \to 6^2P_{3/2}$
λ_1 (nm)	285.29	422.79	852.12
$\Gamma_1/2\pi$ (MHz)	78.8	34.7	5.2
$\mathrm{T_{D,1}}$ (mK)	1.9	0.833	0.125
2^{nd} Trans.	$3^1P_1 \to 3^1D_2$	$4^1P_1 \to 5^1S_0$	$6^2P_{3/2} \to 10^2D_{5/2}$
λ_2 (nm)	880.92	1034.66	563.68
$\Gamma_2/2\pi$ (MHz)	2.0	5.3 [9]	0.49[23]
$\mathrm{T_{D,2}}$ (μK)	48	127	12

second regime is characterized by two-photon processes coupling the ground and the upper state, while the intermediate state is only virtually involved. The dynamics corresponds thus to that of an effective two-level system with the linewidth of the upper state. This process produces a temperature lower than that of the EIT assisted cooling and close to the Doppler limit determined by the lifetime of the upper state.

The theoretical analysis is based on the time evolution of the atomic momentum and the atomic kinetic energy following the model discussed in refs. [19, 20, 21]. We restrict our study to one-dimension, and consider an atom of mass M and momentum p (along the x axis), with internal levels $|0\rangle, |1\rangle, |2\rangle$ with increasing energies $0, \hbar\omega_{01}, \hbar\omega_{02}$, where $|1\rangle$ ($|2\rangle$) decays radiatively into $|0\rangle$ ($|1\rangle$) at rate Γ_1 (Γ_2). The Hamiltonian for the atom interacting with two laser fields at frequencies ω_1, ω_2 and wavevectors k_1, k_2, respectively, is $H_{\rm tot} = H_{\rm at} + V_L + W$, where

$$H_{\rm at} = \frac{p^2}{2M} - \hbar\delta_1|1\rangle\langle 1| - \hbar(\delta_1+\delta_2)|2\rangle\langle 2|,$$
$$V_L = \hbar\Omega_1|1\rangle\langle 0|\cos(k_1x) + \hbar\Omega_2|2\rangle\langle 1|\cos(k_2x) + \text{H.c.},$$

with detunings $\delta_j = \omega_j - \omega_{j0}$ ($j = 1, 2$), Rabi frequencies Ω_1 and Ω_2. W describes the coupling to the modes of the electromagnetic field in the vacuum state. We follow the time evolution of an atom, which at $t = 0$ is in the state $|0, p\rangle$ with energy $E_0(p) = p^2/(2M)$, under the assumption of weak Rabi frequency Ω_1, so that we can treat the coupling of state $|0\rangle$ to state $|1\rangle$ in perturbation theory [22].

We analyse the scattering processes determining changes of atomic momentum. For simplicity, in the following discussion we assume that both lasers are travelling waves. The scattering processes leading to a change of the atomic momentum can be described as consisting of two parts, which add up incoherently:

(i) absorption of one photon with momentum $\hbar k_1$ along the x axis on the transition $|0\rangle \to |1\rangle$ followed by scattering of a photon with momentum $\hbar k_{1s}$. After this process the atom is found in the state $|0, p'\rangle = |0, p + \Delta p_1\rangle$ with $\Delta p_1 = \hbar k_1(1 - \hat\Omega_1 \cdot \hat x)$ where $\hat\Omega_1$ denotes the direction of photon emission. The corresponding scattering rate is $\mathcal{W}_1(p, \hat\Omega_1) = \mathcal{P}_1(\hat\Omega_1)\mathcal{R}_1(p)$, where $\mathcal{P}_1(\hat\Omega_1)$ is the spatial pattern of spontaneous emission for transition $|1\rangle \to |0\rangle$ normalized to unity and the dependence on atomic momentum is given by

$$\mathcal{R}_1(p) = \frac{\Gamma_1\Omega_1^2}{4}\left|\frac{\delta_1' + \delta_2' + \mathrm{i}\Gamma_2/2}{(\delta_1' + \delta_2' + \mathrm{i}\Gamma_2/2)(\delta_1' + \mathrm{i}\Gamma_1/2) - \Omega_2^2/4}\right|^2 \tag{1}$$

where $\delta_1' = -k_1p/m + \delta_1$ and $\delta_2' = -k_2p/m + \delta_2$. Around the two-photon resonance $\delta_1 + \delta_2 = 0$ value the scattering cross section exhibits an asymmetric Fano-like structure as a function of p, typical of interference processes [22]. As in [18], the asymmetry, controlled by δ_1 and Ω_2, affects critically the gradient of the cross section at $p = 0$, and thus the force exerted on the atom at slow velocities.

(ii) The atom undergoes two absorption processes at frequencies ω_1 and ω_2 followed by emission of a photon of wave vector k_{2s} and a second of wave vector k_{1s} such that the atom is pumped back to state $|0\rangle$. After this process the atom is found in the state $|0, p'\rangle = |0, p + \Delta p_2\rangle$ with $\Delta p_2 = \hbar k_1(1 - \hat\Omega_1 \cdot \hat x) + \hbar k_2(1 - \hat\Omega_2 \cdot \hat x)$ where $\hat\Omega_2$ denotes the direction of emission of the k_{2s} photon. The corresponding scattering rate is $\mathcal{W}_2(p, \hat\Omega_1, \hat\Omega_2) = \mathcal{P}_1(\hat\Omega_1)\mathcal{P}_2(\hat\Omega_2)\mathcal{R}_2(p)$. Here $\mathcal{P}_2(\hat\Omega_2)$ is the spatial pattern for spontaneous emission on $|2\rangle \to |1\rangle$ normalized to unity and $\mathcal{R}_2(p)$ is the convolution of transition amplitudes, where energy conservation is imposed on the two-photon emission, and it vanishes when Γ_2 or Ω_2 are set to zero.

The scattering rates allow us to derive the equation for the mean kinetic energy $\langle E\rangle$. A differential change of $\langle E\rangle$ is given by the sum of the energy changes due to each scattering process weighted by their corresponding rates [19]. For cooling based on the atomic excitation by lasers propagating in opposite directions, we consider the absorption-emission paths based on all combinations of copropagating and counterpropagating photons. Taking into the account those contributions, after integrating over the angles of emission, we find

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}\langle E\rangle = \int \mathrm{d}p f(p)\frac{\hbar p}{M}\Big[k_1\left(\mathcal{R}_1(p) - \mathcal{R}_1(-p)\right)\\ + (k_1+k_2)\left(\mathcal{R}_2(p) - \mathcal{R}_2(-p)\right)\Big] + H(\sigma_1, \sigma_2)\end{aligned} \tag{2}$$

where $f(p)$ is the momentum distribution and we have introduced a term describing the heating process

$$\begin{aligned}H(\sigma_1, \sigma_2) = \sigma_1\left(1+\chi_1\right)\frac{\hbar^2k_1^2}{M}\\ + 2\sigma_2\left(\left(1+\chi_1\right)\frac{\hbar^2k_1^2}{M} + \left(1+\chi_2\right)\frac{\hbar^2k_2^2}{M}\right),\end{aligned} \tag{3}$$

with $\chi_i = \int \mathrm{d}\hat\Omega_i\mathcal{P}_i(\hat\Omega_i)(\hat\Omega_i\cdot\hat x)^2$ and $\sigma_i = \int \mathrm{d}pf(p)\mathcal{R}_{i,}(p)$ ($i = 1, 2$). Within Eq. (2) we have neglected a term associated with the correlation in the spontaneous emission of photons on the two atomic transitions. For determining the final temperature we focus on the latest stages of cooling, when the atoms reach their steady state. In this regime we assume $f(p)$ to be gaussian and the Doppler width much less than the natural width of the optical transitions. In this limit, we can expand the rates $\mathcal{R}_i(\pm p) = \mathcal{R}_i(0) \pm \mathcal{R}'_i(0)p$, $\mathcal{R}'_i(0) = d\mathcal{R}_i(p)/dp|_{p=0}$, and $\sigma_i \sim \mathcal{R}_i(0)$. In this limit we obtain

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle E\rangle = -2\alpha\langle E\rangle + H^{\mathrm{o}} \tag{4}$$

where α is the cooling rate

$$\alpha = -2\hbar k_1 \mathcal{R}'_1(0) - 2\hbar\,(k_1 + k_2)\,\mathcal{R}'_2(0) \tag{5}$$

and $H^{\mathrm{o}} = H(\mathcal{R}_1(0), \mathcal{R}_2(0))$ is the heating rate. The temperature T is operationally defined by the relation $k_{\mathrm{B}}T/2 = \langle E\rangle_{\mathrm{st}}$, where k_{B} the Boltzmann constant and $\langle E\rangle_{\mathrm{st}}$, solution of Eq. (4), is the mean energy at steady state. Hence

$$k_{\mathrm{B}}T = \frac{H^{\mathrm{o}}}{\alpha}. \tag{6}$$

The cooling temperature depends through H^{o} on the χ_i parameters determined by the spatial pattern of the spontaneous emission and equal to $2/5$ for a dipole emission. For Doppler cooling on the lower transition the value $\chi_1 = 1$ reproduces the temperature reached in three-dimensional cooling [21].

The evaluation of the final temperature T requires the knowledge of term $\mathcal{R}_2(p)$, which allows for an explicit analytical form only in certain limiting cases. Therefore, in our numerical analysis we have linked $\mathcal{R}_1, \mathcal{R}_2$ to the steady state solution of the Optical Bloch Equations for the atomic density matrix ρ [11]

$$\mathcal{R}_1(p) = \Gamma_1\rho^{\mathrm{st}}_{11}(p) - \Gamma_2\rho^{\mathrm{st}}_{22}(p),\;\; \mathcal{R}_2(p) = \Gamma_2\rho^{\mathrm{st}}_{22}(p). \tag{7}$$

where the steady state populations $\rho^{\mathrm{st}}_{\mathrm{ii}}$ of the three atomic levels depend on the atomic momentum p due to the Doppler effect. The results obtained with this method agree with the prediction obtained using Eq. (1) when Γ_2 (and thus processes described by $\mathcal{R}_2$) is set to zero.

We have analysed cooling temperatures as a function of laser and atomic parameters for the different species listed in Table I. $T_{\mathrm{D}1,2}$ denotes the Doppler cooling limit temperature $\hbar\Gamma_{1,2}/(2k_{\mathrm{B}})$ for a two-level system with the upper lifetime equal to that of the $|1\rangle$ or $|2\rangle$ state, respectively. All optical transitions will be treated as closed ones in order to simplify the theoretical treatment.

The numerical results of Fig. 1(a) show the absorption coefficient of Mg atoms for Doppler cooling on the lower transition (dashed line) and for EIT assisted cooling with a strong near resonant coupling laser on the upper transition (continuous and dotted lines). The coupling laser modifies strongly the absorption profile. The Ω_2 and δ_2 parameters correspond to the experimental conditions of [15]. For the chosen parameters the absorption profile contains a narrow and deep structure at positive or negative δ_1 detunings, depending on the sign of δ_2. In presence of the coupling laser, the cooling rate

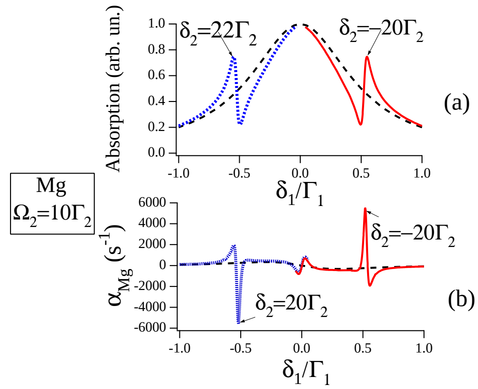

FIG. 1: (color online) Numerical results for the EIT cooling on the Mg transitions of Table I. In (a) absorption coefficient in arbitrary units, and in (b) cooling rate α_{Mg} versus detuning δ_1 at weak intensity of that laser, $\Omega_1 = 0.01\Gamma_1$. Dashed lines for $\Omega_2 = 0$. Continuous (red) line for $\Omega_2 = 10\Gamma_2$ and $\delta_2 = -20\Gamma_2$; dotted (blue) line for $\Omega_2 = 10\Gamma_2$ and $\delta_2 = 20\Gamma_2$.

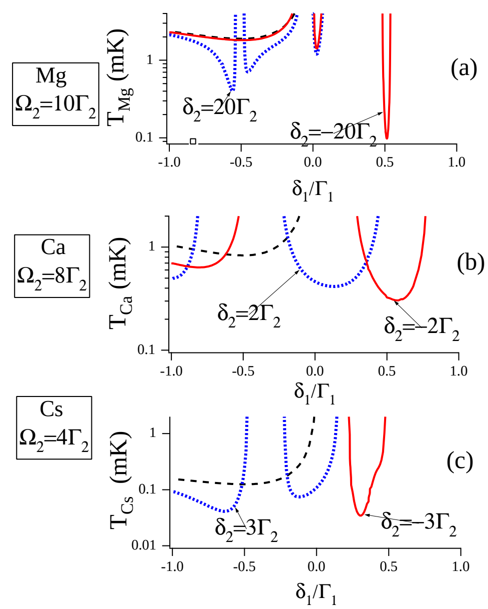

FIG. 2: (color online) Numerical results for the EIT cooling of (a) Mg, (b) Ca, and (c) Cs using the transitions of Table I and supposing $\chi_1 = \chi_2 = 1$ as for a three-dimensional cooling geometry. Temperatures T in mK versus detuning δ_1 at weak intensity $\Omega_1 = 0.01\Gamma_1$, and δ_2, Ω_2 specified values. Dashed line for $\Omega_2 = 0$, i.e., Doppler cooling on the lower transition. Continuous (red) lines for positive values of δ_2. Dotted (blue) lines for negative values of δ_2. The Γ_1 decay rate depends on the atomic species.

α_{Mg}, proportional to the derivative of the absorption coefficient with respect to the frequency, presents deep and narrow structures, as in Fig. 1(b).

Eq. (6) indicates that the α increase of Fig. 1(b) corresponds to a lower cooling temperature, as shown in Fig. 2(a) for the same Mg parameters. However, even if the EIT process increases the cooling rate by twenty times, the lowest temperature is only a factor of five times smaller than the T_{D1} Doppler limit. In fact the cooling temperature is determined by the EIT increase in both α cooling and H^o heating rates. Similar decreases in temperatures are reached for the Ca and Cs transitions as shown in other parts of Fig. 2. The Ca case confirms a peculiar feature of the Mg results: the lowest temperatures are reached using a negative detuning δ_2 and a positive δ_1, corresponding to the largest increase in the damping rate as in Fig. 1(b). For an experimental investigation the positive δ_1 EIT cooling is difficult to realize because a change in the Ω_2 value modifies the δ_1 detuning required for cooling, and in spatial regions of the atomic sample where $\Omega_2 \sim 0$ the Doppler cooling turns into Doppler heating. For Cs the hyperfine structure of the atomic levels produces a sub-Doppler cooling more efficient than the EIT cooling. The interesting point of Cs is that a low temperature is reached even using a laser at short wavelength on the upper transition, corresponding to a large value of k_2 and contributing to an increase the last term in the heating rate of Eq. (3). The term neglected in the heating rate derivation and produced by the correlation in the photon emissions in the upper and lower transitions has an amplitude similar to those containing the $\chi_{\rm i}$ parameters. Therefore it would increase the temperature by less than fifty percent. We have not performed an accurate optimization of all laser parameters. However for the explored parameters of Mg and Ca the temperatures reached in this EIT cooling are not very close to the Doppler cooling limit on the upper transition.

In the two-photon cooling the intermediate state $|1\rangle$

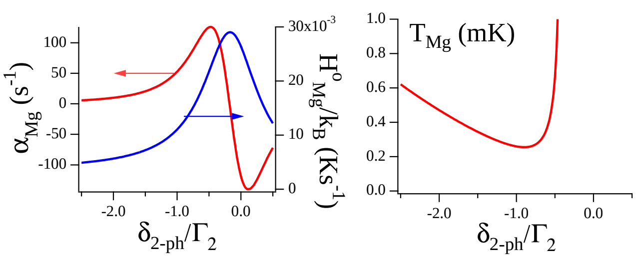

FIG. 3: (color online) Results for Mg two-photon cooling. Cooling rate $\alpha_{\rm Mg}$ and heating rate $H^o_{\rm Mg}$ divided by $k_{\rm B}$ (left), temperature $T_{\rm Mg}$ (rigth), in units of $T_{\rm D_2}$, versus the 2-photon detuning $\delta_{2-\rm ph} = \delta_1 + \delta_2$ of the coupling laser. Parameters: $\Omega_1 = 0.0\Gamma_1$, $\delta_1 = -40\Gamma_1$ and $\Omega_2 = 50\Gamma_2$.

does not play a role and an effective two-level system composed by the states $|0\rangle$ and $|2\rangle$ should produce a temperature close to the Doppler limit $T_{\rm D2}$ on the upper level. This is confirmed by the numerical analysis of Fig. 3 for Mg, where the ω_1 laser is far detuned by 40 linewidths from the $|0\rangle \rightarrow |1\rangle$ transition and the frequency sum $\delta_{2-\rm ph} = \delta_1 + \delta_2$ is scanned over a small frequency interval determined by the Γ_2 decay rate. Temperatures close to the $T_{\rm D2}$ limit and much smaller than $T_{\rm D1}$ are obtained for different values of δ_1 and Ω_2. Similar results are obtained for the Ca and Cs transitions. The main difficulty in applying this scheme is that the difference frequency $\delta_1 - \delta_2$ should controlled with a Γ_2 precision. That is difficult in the case of an ultraviolet laser as required for Mg.

An additional difference between the EIT and two-photon cooling schemes is their $\Delta v_{\rm c}$ velocity capture range. The capture range associated with the EIT process is derived from the data of Fig. 1 for the frequency range where the the damping rate is significantly different from that of the Doppler cooling process. We obtain $\Delta v_{\rm c} \sim 0.2\Gamma_1/k_1$, to be compared to the value of Γ_1/k_1 associated with the Doppler cooling. For the two-photon cooling the velocity capture range is reduced to $\Delta v_{\rm c} \sim \Gamma_2/k_2$ as demonstrated by the data of Fig. 3. Therefore a lower temperature is reached at the expense of the lower efficiency in the collection of the cooled atoms, a behaviour similar to that of sub Doppler cooling.

In conclusion, in laser cooling based on two-color excitation of an atomic cascade, the basic phenomenon is the mixing of the intermediate state with an upper state having a longer lifetime. This mixing scheme is complementary to that of quenching cooling where the role of the mixed states is exchanged. We have examined the limiting cases of EIT-assisted and two-photon cooling. For an experimental investigation a lower cooling may be reached using two-photon excitations with the intermediate state excited only virtually. In the EIT assisted scheme, a larger capture range and a more flexible combination with the single photon Doppler cooling enhance the cooling performance.

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